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Technical Report No. 19

Electronic and Ionic Transport in Processable Conducting Polymers

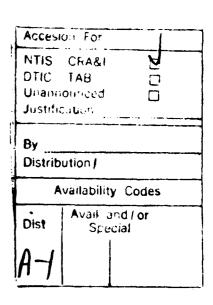
by

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John R. Reynolds
Krishnan Rajeshwar
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April 10, 1990





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Description of Progress

We have prepared 16 bis(2-heterocycle)benzenes using thiophene, furan and pyrrole as the heterocycle. These precursor monomers for the preparation of a new family of conjugated processable polymers are substituted at the 2 and 5 positions of the phenylene ring with methyl, hexyl, methoxy, heptoxy and dodecyloxy substituents to yield both symmetric and unsymmetric molecules. Oxidative polymerization has been accomplished using both chemical (FeCl₃) and electrochemical methods. In the case of the 2-thienyl derivatives, the highest conductivity has been observed for the NOPF₆ doped dimethoxy substituted polymer (2 Ω^{-1} cm⁻¹). The dimethyl substituted polymers, however, exhibit significantly lower conductivity (10⁻⁶ Ω^{-1} cm⁻¹). Correlations of the electronic properties of the polymers have been made with theoretically modeled minimum energy conformations and extended Hückel determined band structures. A benefit of the symmetrically substituted polymers is that they contain no configurational isomerism in the form of head-head defects. This is a common problem with the poly(3-alkylthiophenes).

Molecular weight studies of poly(3-alkylthiophenes) using GPC in conjunction with our multiangle laser light scattering (MALLS) detector (which gives absolute molecular weights) has shown that weight average molecular weights for the series where alkyl = C_4H_9 , C_6H_{13} , C_8H_{17} , $C_{10}H_{21}$ and $C_{12}H_{25}$ varied from 204,000-398,000 and the molecular weight at the maximum in concentration varied from 88,000 to 320,000. These single values at the maximum are the *absolute* molecular weights. We have also prepared several different samples of poly(3-octylthiophene) using FeCl₃, and have shown that there is a variation in the molecular weights from sample to sample. Thus the weight average molecular weights ranged from 113,000 to 307,000 while the molecular weight at the peak maximum ranged from 73,000 to 160,000. Fibers of blends of poly(3-decylthiophene) with low density polyethylene (40:60 and 20:80) have been prepared in our continuing collaboration with Hoechst-Celanese Research Division, Summit, New Jersey. Electron microscopy has shown that these blend fibers are phase separated and are actually composites of poly(3-decylthiophene) in a matrix of polyethylene. EDAX studies have confirmed this.

Following our initial observations of the ion exchange properties of polypyrrole, we have recently completed quantitative studies on the ion binding properties of "self-doped" poly(pyrrole propanesulfonate) thin films. These studies show that these films can tenaciously bind useful photophysical molecules such as $Ru(bpy)_3^{2+}$ (bpy = 2,2'-bipyridyl). Efforts to extend these findings to photoelectrochemical applications related to solar energy storage are underway.

We have also demonstrated how *in situ* surface photochemistry at chemically modified electrodes offers a powerful route to catalyst generation at the surface. One such "molecularly engineered" system comprises the synthesis of Prussian blue from surface-immobilized ferriferrocyanide reaction centers at a quarternized polyvinylpyridine-modified glassy carbon surface. Aside from the intrinsic advantages related to the high *local* concentration (several moles/liter) at chemically modified surfaces, these new heterogeneous (essentially two-dimensional) surfaces offer useful insights and comparisons with the chemistry of their homogeneous (3-D) solution counterparts.

Publications

Papers Published

Qiu, Y-J. and Reynolds, J. R. "Poly[3,6-(carbaz-9-yl)propanesulfonate]: A Self-Doped Polymer with Both Cation and Anion Exchange Properties," J. Electrochem. Soc. 1990, 137, 900-904.

Sharma, S. C., Krishnamoorthy, S., Naidu, S. V., Eom, C. I., Krichene, S., and Reynolds, J. R. "Positron Annihilation and Conductivity Correlations in Poly(pyrrole tosylate) and Poly(pyrrole fluoride)," *Phys. Rev. B.* 1990, 41, 5258-5265.

Shaffer, T. D. and Kramer, M. C., "Cyclization versus Polymerization in Phase Transfer Catalyzed Polythioetherification," *Makromol. Chem.* 1990, 91, 71-79.

Papers in Press

Nayak, K. and Marynick, D. S. "The Interplay Between the Geometric and Electronic Structures of Polyisothianaphthene and Polyisonaphthothiophene, Polythieno(3,4-b)pyrazine and Polythieno(3,4-b)quinoxaline," *Macromolecules*, in press.

Gieselman, M. G. and Reynolds, J. R. "Poly(p-phenyleneterephthalamide propanesulfonate): A New Polyelectrolyte for Application to Conducting Molecular Composites," *Macromolecules*, in press.

Wang, F. and Reynolds, J. R. "Soluble and Electroactive Nickel Bis(dithiolene) Polymers Prepared Via Metal Complexation Polymerization," *Macromolecules*, in press.

Reynolds, J. R. and Pomerantz, M. "Processable Electronically Conducting Polymers," in "Electroresponsive Molecular and Polymeric Systems," Skotheim, T. A., Ed.; Marcel Dekker: New York; Vol. 2, in press.

Basak, S., Rajeshwar, K., and Kaneko, M. "Ion Binding of Poly{pyrrole-co[3-(pyrrol-1-yl)-propanesulfonate]} Thin Films," *Anal. Chem.*, in press.

Papers Submitted for Publication

Reynolds, J. R., Ruiz, J. P., Child, A. D., Marynick, D. S., and Nayak, K. "Electrically Conducting Polymers Containing Alternating Substituted Phenylenes and Bithiophene Repeat Units," *Macromolecules*, submitted for publication.

Basak, S., Rajeshwar, K., and Kaneko, M. "In Situ Photogeneration of a Catalyst on a Chemically Modified Electrode Surface: Application to a Mixed Valent Hexacyanoferrate System," J. Chem. Soc., Chem. Commun., submitted for publication.

Shaffer, T. D. and Sheth, K. A. "Mesomorphic Polyazomethine Ethers Containing Dibenzo-18-crown-6 Units," *Makromol. Chem., Rapid Commun.*, submitted for publication.

Meetings Attended and Talks Presented

John Reynolds attended the Gordon Research Conference on Polymer Chemistry, Ventura, California, January, 1990, and presented a paper entitled "Metal Complex Polymers."

John Reynolds attended the 17th Annual Water-Borne and Higher-Solids Coatings Symposium on Advances in Polymer Synthesis, New Orleans, Louisiana, February, 1990, and presented a paper entitled "Electropolymerization in the Synthesis of Conducting Materials."

Krishnan Rajeshwar presented an invited seminar entitled "Charge Transport/Storage in Biconductive Oxides and Polymers" to the Chemistry Department at Texas A&M University, College Station, Texas, March, 1990.

Visitors to UTA

Professor Allan S. Hoffman of the University of Washington presented a seminar at U. T. Arlington on March 26, 1990, entitled "Biomedical Applications of Plasma Polymerization: Two Case Studies: The Small Diameter Vascular Graft and Non-Fouling Surfaces," detailing his program in the surface properties of polymers and their biomedical applications.

New Collaboration with a DoD Laboratory

A collaborative, funded program was initiated with researchers at the Naval Weapons Center (Dr. Geoff Lindsay) entitled "Synthesis of Electrochemically Switchable Conducting Polymer Films."

New Personnel

Dr. J. R. Dharia joined our group as a postdoctoral fellow.